Dissipative binding of atoms by non-conservative forces

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The formation of molecules and supramolecular structures results from bonding by conservative forces acting among electrons and nuclei and giving rise to equilibrium configurations defined by minima of the interaction potential. Here we show that bonding can also occur by the non-conservative forces responsible for interaction-induced coherent population trapping. The bound state arises in a dissipative process and manifests itself as a stationary state at a preordained interatomic distance. Remarkably, such a dissipative bonding is present even when the interactions among the atoms are purely repulsive. The dissipative bound states can be created and studied spectroscopically in present-day experiments with ultracold atoms or molecules and can potentially serve for cooling strongly interacting quantum gases.

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In most experiments investigating coherent quantum dynamics, dissipation is an undesirable process. However, there have been several recent theoretical proposals for turning controlled dissipation into a useful resource, e.g., for the realization of interesting many-body quantum states [1–3]. The main idea is to engineer the interaction with the environment such that the combination of coherent and dissipative dynamics drives the system in question to a stationary state identical to the quantum state of interest. The feasibility of such a reservoir engineering has already been demonstrated experimentally [4].

In our work, we employ a similar idea to achieve a dissipative bond between two interacting atoms (or molecules), i.e., a stationary state of the scattering dynamics that confines the atoms at a fixed distance. Our results thus allow to extend the notion of a bonding mechanism from purely conservative to dissipative forces. Owing to the unprecedented control available for ultracold atoms and molecules [5], the characteristics of the resulting molecules such as bond lengths and spectroscopic properties are highly tunable.

As a specific physical implementation, we first consider two interacting atoms whose motion is restricted to one spatial dimension, see Fig. 1 (a). For ultracold atoms and molecules, such a constraint can be enforced by appropriate trapping potentials [5]. We assume the internal level structure of the atoms as shown in Fig. 1 (b): each has one internal state $|1\rangle$ exhibiting a distance-dependent interaction shift, and two other noninteracting states $|2\rangle$ and $|3\rangle$, with $|2\rangle$ undergoing spontaneous decay into states $|1\rangle$ and $|3\rangle$. This particular level structure is quite common in ultracold atoms and molecules, here, we focus on two different possible realizations based on Rydberg-dressed atoms [6–8] or laser-cooled molecules [9–11]. While our approach is general, we focus on dipole-dipole interactions, as this brings in additional tunability due to the anisotropy of the interaction potential [12]. We generalize our results to higher spatial dimensions and discuss the prospects of using dissipative bonding as a cooling mechanism for strongly interacting quantum gases.

The process behind the formation of the bond is coherent population trapping (CPT) [13, 14]. In our setup, we include two counter-propagating laser beams; the one corresponding to Rabi frequency Ω_- is resonant with the transition between states $|2\rangle$ and $|3\rangle$, and the one with Rabi frequency Ω_+ is detuned by Δ from the resonance

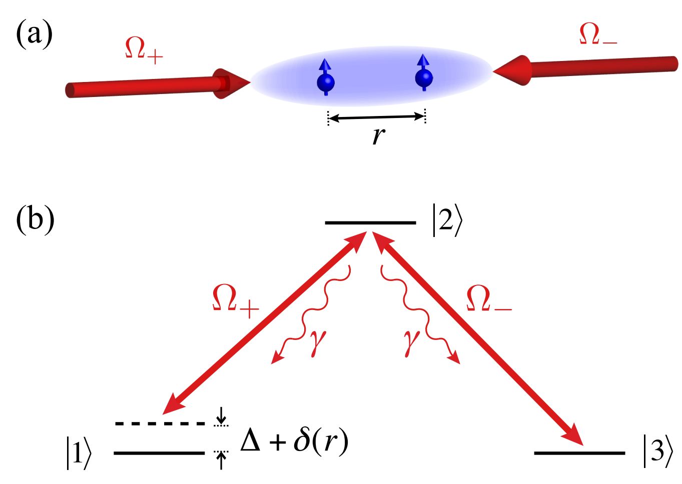

FIG. 1: Setup of the system. (a) Strong confinement by external fields restricts the atoms' motion to one spatial dimension parallel to the two counter-propagating laser beams represented by the driving fields with Rabi frequencies $\Omega_\pm$; (b) Internal level structure of the atoms. Two internal states $|1\rangle$ and $|3\rangle$ are coupled to a metastable state $|2\rangle$ spontaneously decaying with a rate γ. In the non-interacting limit the field Ω_+ is detuned by Δ from the $|1\rangle - |3\rangle$ transition; state $|1\rangle$ is subject to a dipolar interaction shift $\delta(r)$ which depends on the interparticle distance r; $|2\rangle$ and $|3\rangle$ are noninteracting.

between $|1\rangle$ and $|3\rangle$. CPT entails a dark state, i.e., a stationary state which cannot absorb photons.

For a system of two atoms subject to the electromagnetic field treated as a reservoir [15], the dynamics is obtained from the quantum master equation,

$$\frac{d\rho}{dt} = -i\hbar\,[H,\rho] + \sum_n \gamma_n \left(c_n \rho c_n^\dagger - \frac{1}{2}\{c_n^\dagger c_n, \rho\} \right), \quad (1)$$

whose stationary solution is given by the condition $\mathrm{d}\rho/\mathrm{d}t = 0$. Here, H is the Hamiltonian describing the two atoms and their interaction with the laser fields, ρ is the density operator describing the two atoms, and γ_n and c_n are the rates and corresponding jump operators associated with spontaneous decay. The coherent part of the dynamics according to the quantum master equation (1) is given by the two-atom Hamiltonian,

$$\begin{aligned} H = \sum_{k,i} \Bigg[& \frac{\hbar^2 k^2}{2m} |k\rangle\langle k|_i - \frac{\Omega_-}{2} \left(|1, k+\Delta k\rangle\langle 2, k|_i + \text{h.c.} \right) \\ & - \frac{\Omega_+}{2} \left(|3, k-\Delta k\rangle\langle 2, k|_i + \text{h.c.} \right) - \Delta |1,k\rangle\langle 1,k|_i \Bigg] \\ & + \sum_{k,k',q} \tilde{V}_{dd}(q) |1, k-q\rangle_1 |1, k'+q\rangle_2 \langle 1,k|_1 \langle 1,k'|_2. \end{aligned} \quad (2)$$

Here, $i = 1,2$ and k label the atoms and their corresponding momentum states, and $\tilde{V}_{dd}(q)$ is the Fourier transform of the dipole-dipole interaction potential. The dissipative part of Eq. (1) contains the rates $\gamma_n = \gamma$ and jump operators $c_n = \sum_k |k+\Delta k_n, j_n\rangle\langle 2,k|_{i_n}$ in the Lindblad form, responsible for the decay of each atom from state $|2\rangle$. The index $i_n = 1,2$ runs over the two atoms, while $j_n = 1,3$ accounts for the two final states, and Δk_n contains all possible values of the emitted photon's wave vector [16].

From the master equation (1), one can extract an effective non-Hermitian Hamiltonian, $H_{\text{eff}} = H - iV_d$, containing a dissipative potential,

$$V_d = \hbar \sum_n \frac{\gamma_n}{2} c_n^\dagger c_n. \quad (3)$$

The zero-energy eigenstate of H_{eff} corresponds to the dark state $|\psi_{\text{dark}}\rangle$ if its dissipation vanishes, i.e., $\langle c_n^\dagger c_n\rangle = 0$. In the non-interacting but resonant case, $\delta(r) \equiv \Delta = 0$, the dark state for each atom is given by $(|1\rangle - |3\rangle)/\sqrt{2}$. Taking the coupling to translation via the Doppler effect into account retains this dark state and gives rise to the well-known velocity-selective CPT [14]. In contrast, here we consider the regime where the kinetic energy is small compared to the dipolar interaction between the atoms, which in turn is small compared to both the laser driving and the decay, i.e., $(\hbar k)^2/2m \ll \Delta, \delta(r) \ll \gamma, \Omega_\pm$. In the limit of infinite mass m, we find via perturbation theory that the dissipation has a minimum when the atoms are separated by a distance r_d where the dipolar interaction $\delta(r)$ destructively interferes with the detuning Δ the most,

$$r_d = \left[\frac{d^2}{4\pi\varepsilon_0 \hbar \Delta} \left(\frac{5}{8} + \frac{\Omega_\pm^2}{4\gamma^2} \right) \right]^{1/3}. \quad (4)$$

Here d is the dipole moment of state $|1\rangle$, ε_0 the vacuum permittivity, and $\hbar$ Planck's constant. While there is still some residual dissipation for atoms confined to the vicinity of r_d, the probability to find the atoms separated by r_d is strongly enhanced. Such a confinement of the atoms amounts to the formation of a dissipative bond. Additional perturbations arising due to a finite mass lead to a stationary state that exhibits a distribution of distances sharply centered around r_d rather than a single fixed distance.

We now turn to numerical simulations to confirm the existence of the dissipative bond. We solve the quantum master equation, Eq. (1), using the wave-function Monte-Carlo method [16]. During the time evolution the atomic positions are redistributed by the kinetic energy and photon recoil, and the atomic population is accumulated in the vicinity of r_d on a fast timescale of a few tens of γ^{-1}. For an ensemble of atoms with randomly distributed initial conditions we observe the appearance of a quasi-stationary distribution strongly peaked around r_d, see Fig. 2. The results presented in Fig. 2 correspond to a state after the evolution time of 75 γ^{-1}, averaged over 250 realizations with random initial conditions. In each realization two atoms are confined in a one-dimensional box $L \sim 6r_d$, which corresponds to the three-dimensional particle density of 3×10^{11} cm^{-3}.

For even longer times the spatial distribution of the atoms appears to evolve towards a completely delocalized equal-weight superposition of positions compatible with a relative distance $r_d \approx 500\,\text{nm}$. This constitutes evidence that in the long-time limit the state corresponding to the dissipative bond is a nearly pure state with a low entropy. Once the dissipative bond has been formed, it is possible to further reduce the probability of subsequent spontaneous decay events by decreasing the Rabi frequencies $\Omega_\pm$. This leads to an effective decay rate $\gamma_{\text{eff}} = \gamma p_2$, with p_2 the total probability of any of the atoms being in state $|2\rangle$. We find that the reduction of the Rabi frequencies does not lead to a significant increase of the spread about r_d, and that lifetimes of the bound state exceeding $\gamma_{\text{eff}}^{-1} \approx 0.1\,\text{s}$ can be achieved.

Experimental observation of the resulting bound states can be performed by making use of the techniques well-established in the area of ultracold quantum gases. In particular, a suitable setting would be a many-body system, in which the atoms are essentially uncorrelated at the beginning, with an average interatomic distance much larger than r_d, so that many-body effects could be neglected. Then, the existence of the bound state will

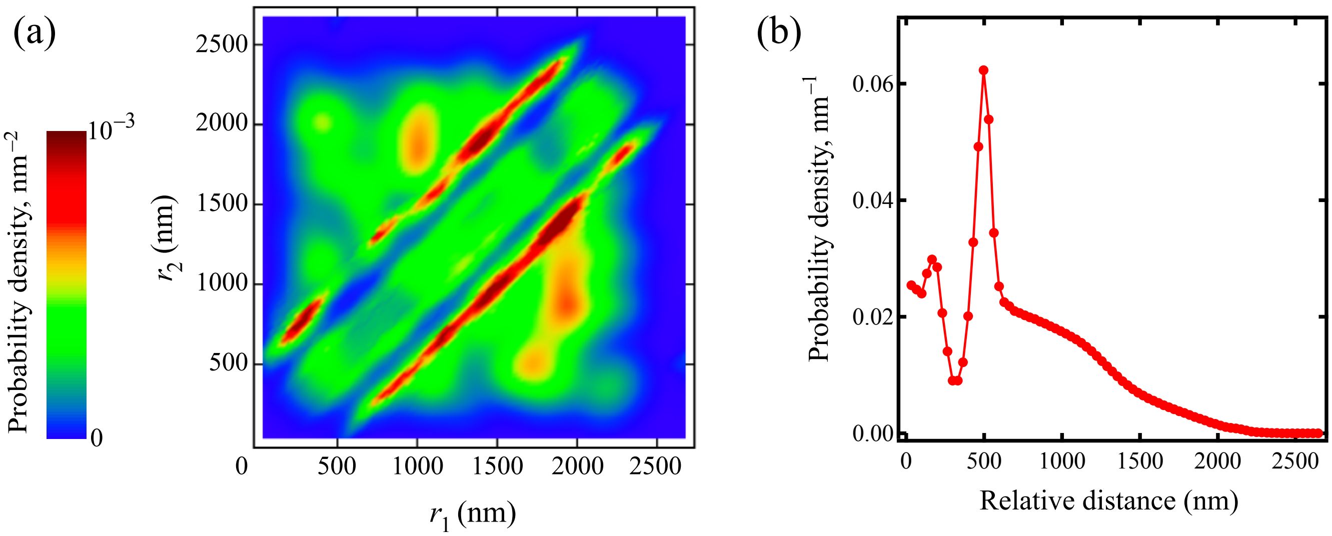

FIG. 2: Probability density distributions indicating the formation of the dissipatively bound states: (a) the absolute coordinates of the two particles, r_1, r_2, are delocalized; (b) the projection onto the relative distance $r = |r_1 - r_2|$ exhibits a sharp peak around $r_d = 500\,\text{nm}$. Parameters correspond to a pair of Rydberg-dressed Cesium atoms (see text).

appear as a sharp peak in the pair-correlation function, see Fig. 2 (b), which is readily accessible through Bragg scattering [17] or noise correlation spectroscopy [18, 19].

The above results can be generalized to higher spatial dimensions, thereby allowing to study the motion of the bound atoms. This merely requires the inclusion of additional counter-propagating laser beams as used in conventional laser cooling setups [20]. For a two-dimensional configuration, we consider the case when the quantization axis defined by the electric dipole moment is perpendicular to the plane of the motion, i.e., when the dipolar interaction is always repulsive. Then, the dissipative bond restricts the relative radial motion to a ring where the atoms are separated by r_d. The effective Hamiltonian $H_{\rm eff}$ for the remaining angular motion is then described by a two-dimensional rigid rotor, $H_{\rm eff} = BJ_z^2$, with J_z the projection of the angular momentum on the quantization axis. The rotational constant $B = \hbar^2/(mr_d^2)$ can be widely tuned by changing r_d; for typical experimental realizations, we find that values of $B \sim 100 - 1000\,\text{Hz}$ are realistic, i.e., by far exceeding the effective decay rate $\gamma_{\rm eff}$. The rotational spectrum of the bound atoms can be then probed with standard techniques of molecular spectroscopy.

In three spatial dimensions, the constrained relative motion of the two atoms is even richer due to the anisotropy of the dipolar interaction. The dynamics is confined to a surface defined by

$$r^3 = r_d^3(1 - 3\cos^2\vartheta), \tag{5}$$

with ϑ the angle between the collinear dipoles and the interatomic radius-vector. Due to the rotational symmetry about the quantization axis, the projection of the angular momentum, J_z, is conserved, allowing to reduce the problem to an effective one-dimensional problem. The effective potential corresponds to an azimuthally-symmetric double well centered at $\vartheta = \pi/2$, resulting in an almost equidistant spectrum consisting of tunneling doublets, Fig. 3 (a), similar to those encountered in microwave spectra of ammonia [21] or molecules in strong laser fields [22]. Fig. 3 (b) features the probability density distributions on the three-dimensional surface. Although the results shown in Fig. 3 correspond to $J_z = 0$, energy spectra and wavefunctions for other values of J_z exhibit similar features.

Experimental implementation.— We consider two different realizations of our setup depicted in Fig. 1, based on Rydberg-dressed Cesium atoms and laser-cooled molecules. In the case of Rydberg dressing [6–8], the states $|1\rangle$ and $|3\rangle$ are chosen as different hyperfine components of the electronic ground state, $6^2S_{1/2}$, of Cesium. The state $|1\rangle$ is provided with a dipole moment of $d = 15\,\text{D}$ perpendicular to the 1D trap due to Rydberg dressing in an external electric field; laser fields $\Omega_\pm = \gamma/2$ drive the $6^2S_{1/2} \to 6^2P_{3/2}$ transition having a linewidth of $\gamma = 2\pi \times 5.2$ MHz. We would like to stress that in contrast to the previous Rydberg dressing proposals for the observation of interactions in a Bose-Einstein condensate, the requirements on coherence times for the observation of the dissipative bond are much less stringent; preserving coherence on the timescale of the rotational constant $B \sim 100\,\text{Hz}$ can be achieved in present-day experiments [23–28], see Ref. [29] for a recent review.

Although we exemplified our scheme by the case of bonding between a pair of atoms, it is equally applicable

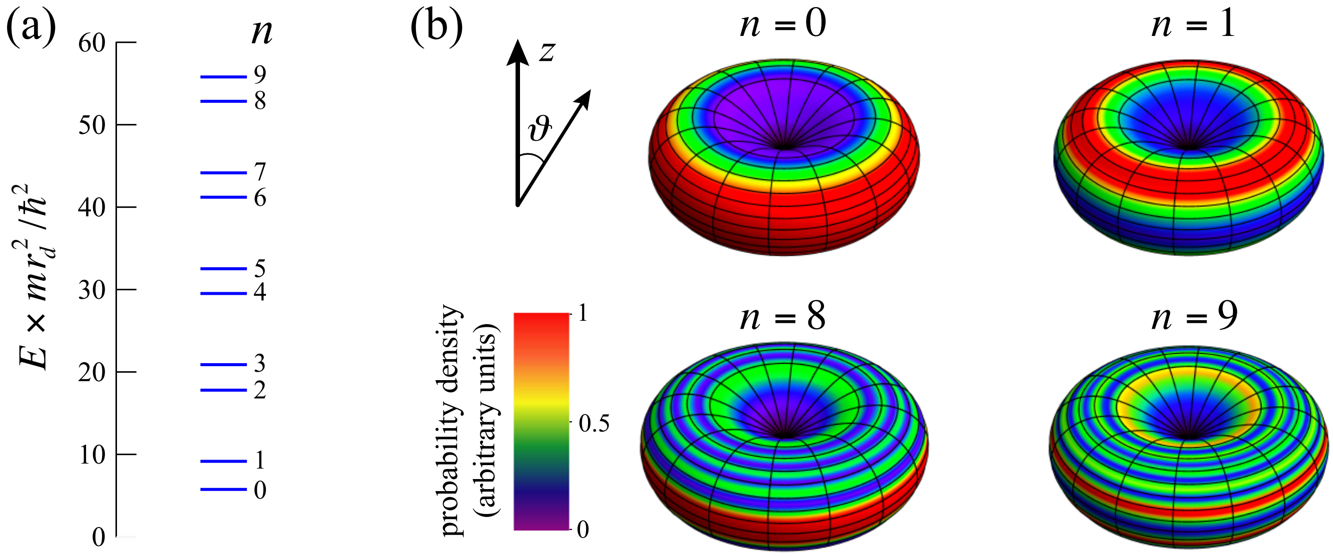

FIG. 3: Three-dimensional eigenstates ($J_z = 0$): (a) lowest energy levels featuring tunneling doublets due to the azimuthal symmetry of the interaction potential; (b) probability density distributions. The angle ϑ gives the orientation of the collinear dipoles with respect to the vector connecting them. The quantum number n gives the number of nodes of the probability density distribution (purple color) depending on the angle ϑ.

to bonding between a pair of molecules, such as strontium monofluoride (SrF) that is being laser-cooled [10]. For SrF, different hyperfine components, $F = 0, 1$, of the $X^2\Sigma^+(v = 0; N = 1, J = 1/2)$ state are chosen as $|1\rangle$ and $|3\rangle$. Here, a dipole moment of 3.5 D in the rotating frame is imprinted on $|1\rangle$ via microwave dressing [30], while the fields $\Omega_\pm = \gamma/2$ drive transitions to the electronically excited $A^2\Pi_{1/2}(v' = 0; N = 0, J' = 1/2)$ whose natural linewidth is $\gamma = 2\pi \times 7$ MHz.

Finally, we note that the mechanism underlying the formation of the dissipative bond could be used for cooling of strongly-interacting many-body systems. Clearly, the appearance of a low entropy stationary state independent of the initial conditions already amounts to a demonstration of strong cooling. In the high-density regime, the correlation length r_d will be modified by an effective coordination number, embodying the interactions of an individual atom with its surroundings in the stationary state. Despite this renormalization, the formation of dissipatively bound complexes [31] in free space or even the realization of dipolar crystals [32, 33] appear possible.

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